

# Surface ODEs in the polar regions

IASOA Ozone WG  
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# Outline

## 1. Brief review of surface ODEs

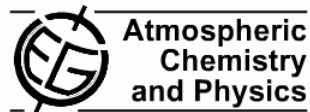
- overview
- sources of halides
- issues for lab, model, and observation

## 2. Few highlights of recent case studies

## 3. Challenges & opportunities for observation group

# Reviews of surface ODEs

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## Halogens and their role in polar boundary-layer ozone depletion

W. R. Simpson<sup>1</sup>, R. von Glasow<sup>2</sup>, K. Riedel<sup>3</sup>, P. Anderson<sup>4</sup>, P. Ariya<sup>5</sup>, J. Bottenheim<sup>6</sup>, J. Burrows<sup>7</sup>, L. J. Carpenter<sup>8</sup>, U. Frieß<sup>9</sup>, M. E. Goodsite<sup>10</sup>, D. Heard<sup>11</sup>, M. Hutterli<sup>4</sup>, H.-W. Jacobi<sup>17</sup>, L. Kaleschke<sup>12</sup>, B. Neff<sup>13</sup>, J. Plane<sup>11</sup>, U. Platt<sup>9</sup>, A. Richter<sup>7</sup>, H. Roscoe<sup>4</sup>, R. Sander<sup>14</sup>, P. Shepson<sup>15</sup>, J. Sodeau<sup>16</sup>, A. Steffen<sup>6</sup>, T. Wagner<sup>9,14</sup>, and E. Wolff<sup>4</sup>

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## Halogen activation via interactions with environmental ice and snow in the polar lower troposphere and other regions

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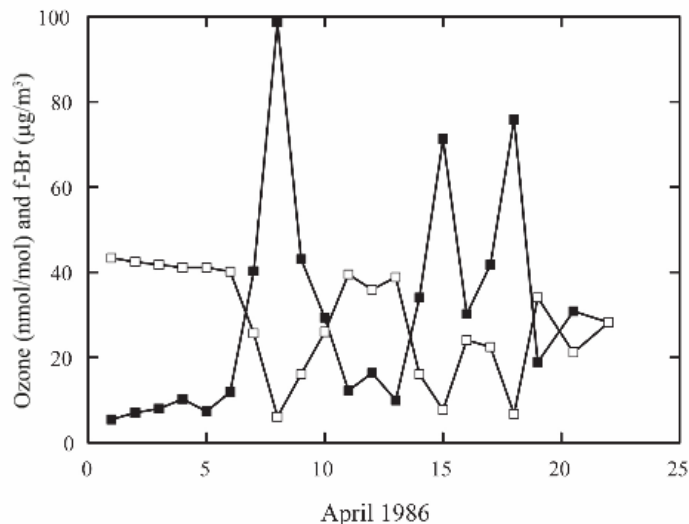
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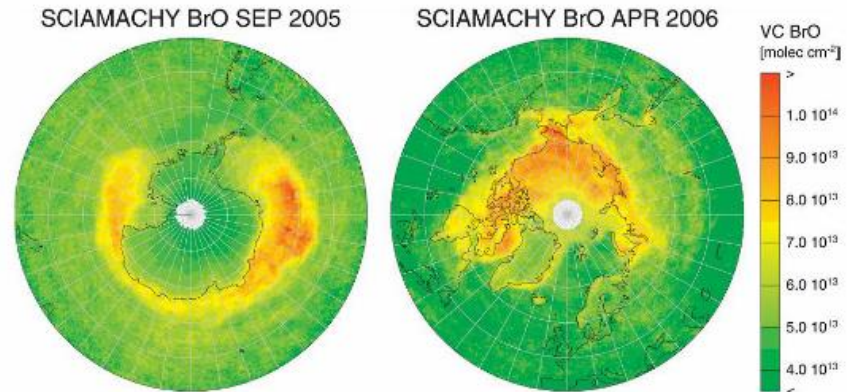
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# Overview of surface ODEs



**Fig. 2.** The first published observation of the anticorrelation between ozone and filterable bromine, measured at Alert. Filled squares show filterable Br (f-Br) in  $\text{ng}/\text{m}^3$ , from 24 h filter pack, open squares show daily averaged ozone in nmol/mol (ppbV). Reprinted by permission from Macmillan Publishers LTD: Nature, Barrie et al. (1988), copyright 1988.

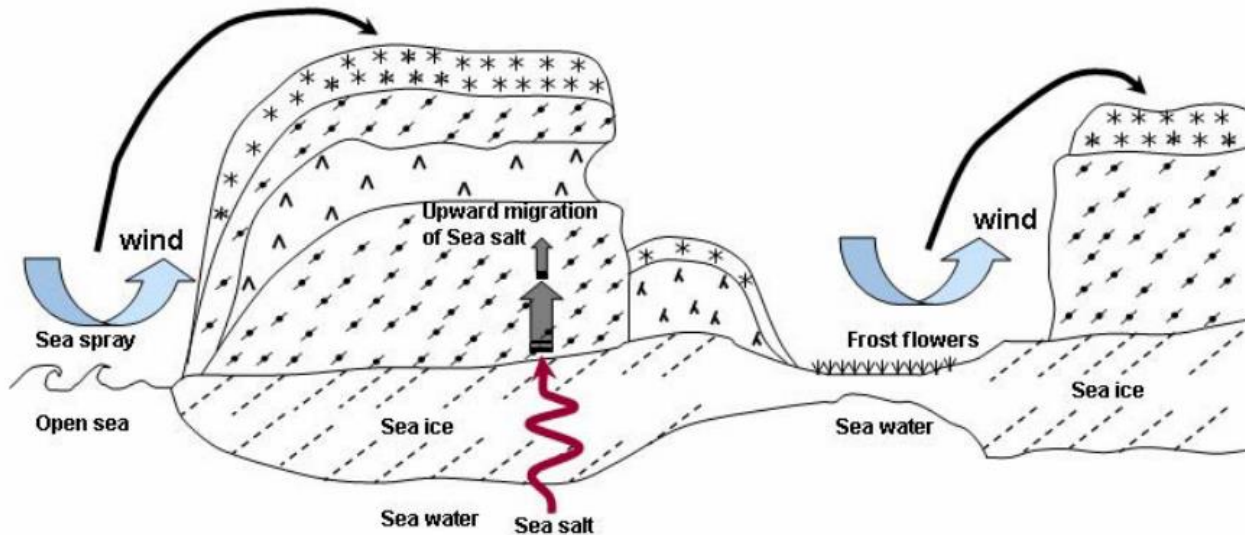


**Fig. 9.** Vertical columns for BrO for both hemispheres in spring. Data from SCIAMACHY, courtesy of A. Richter.

- Polar spring
- Depleted surface ozone
- Increased BrO/halogen

Simpson et al., 2007

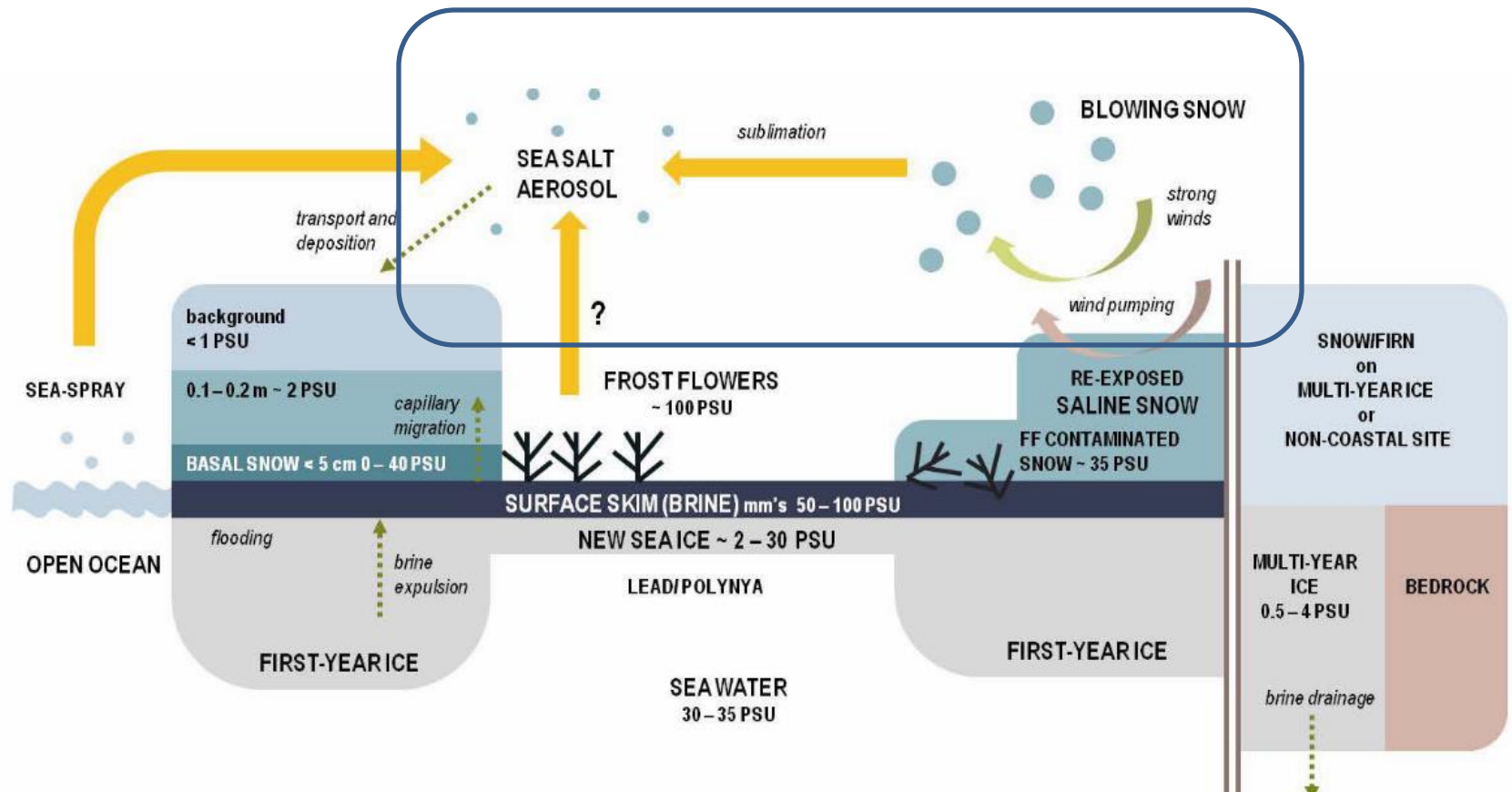
# Sources of halides in the polar boundary layer



**Fig. 14.** Illustration of the main three processes suspected of supplying sea salt ions to marine snow: wind-transport of sea spray, upward migration from sea ice, and wind-blown frost flowers. Reprinted from Dominé et al. (2004) with the permission of the authors.

Simpson et al., 2007

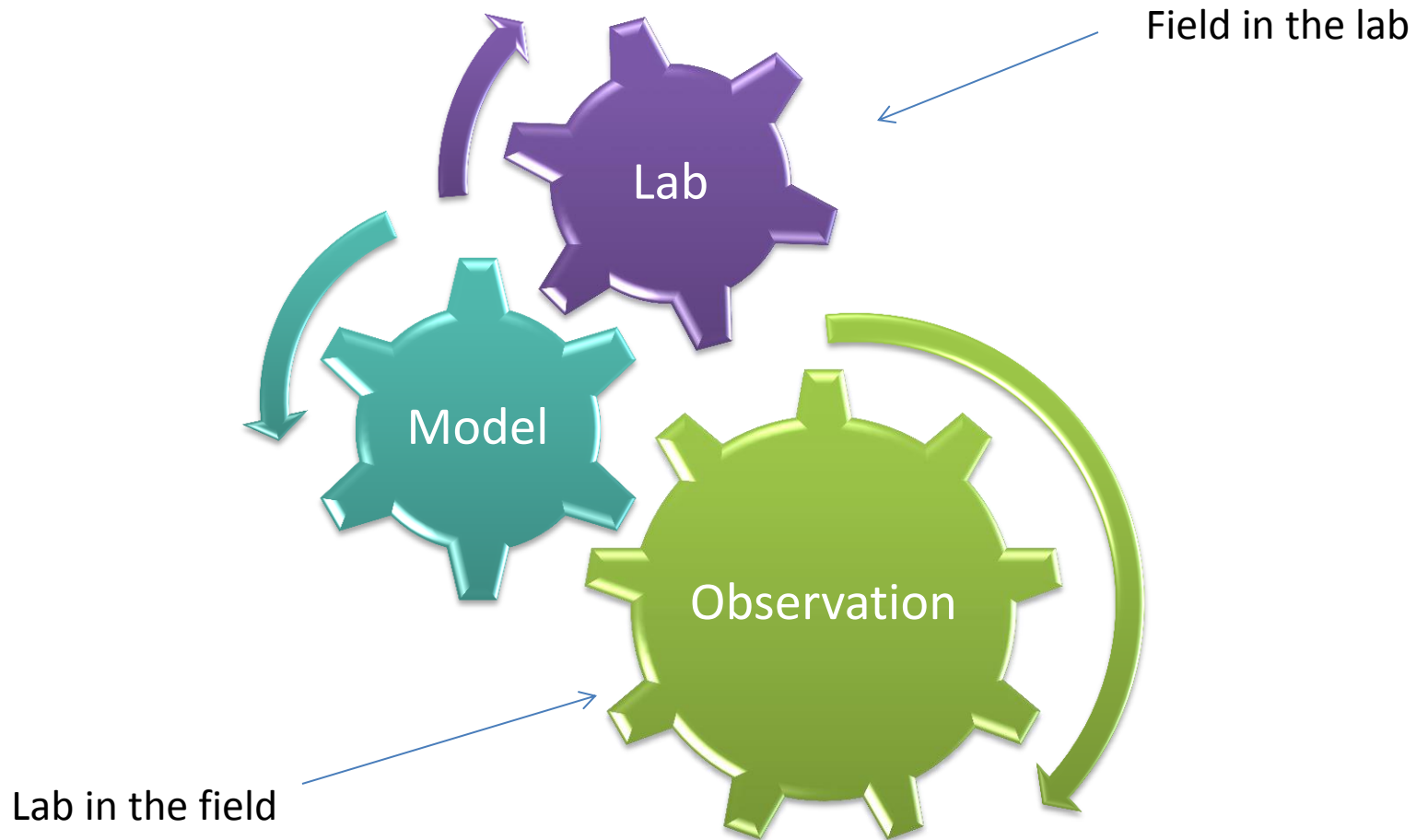
# Sources of halides in the polar boundary layer



**Fig. 1.** Sources of halides in the polar boundary layer. A range of salinity values is indicated: frost flowers (Rankin et al., 2002), brine, new ice (Ehn et al., 2007), multi-year ice (Timco and Weeks, 2010), basal snow (Toyota et al., 2011b), snow < 0.2 m (Toyota et al., 2011b), background snow (Massom et al., 2001), frost flower (FF) contaminated snow (Obbard et al., 2009).

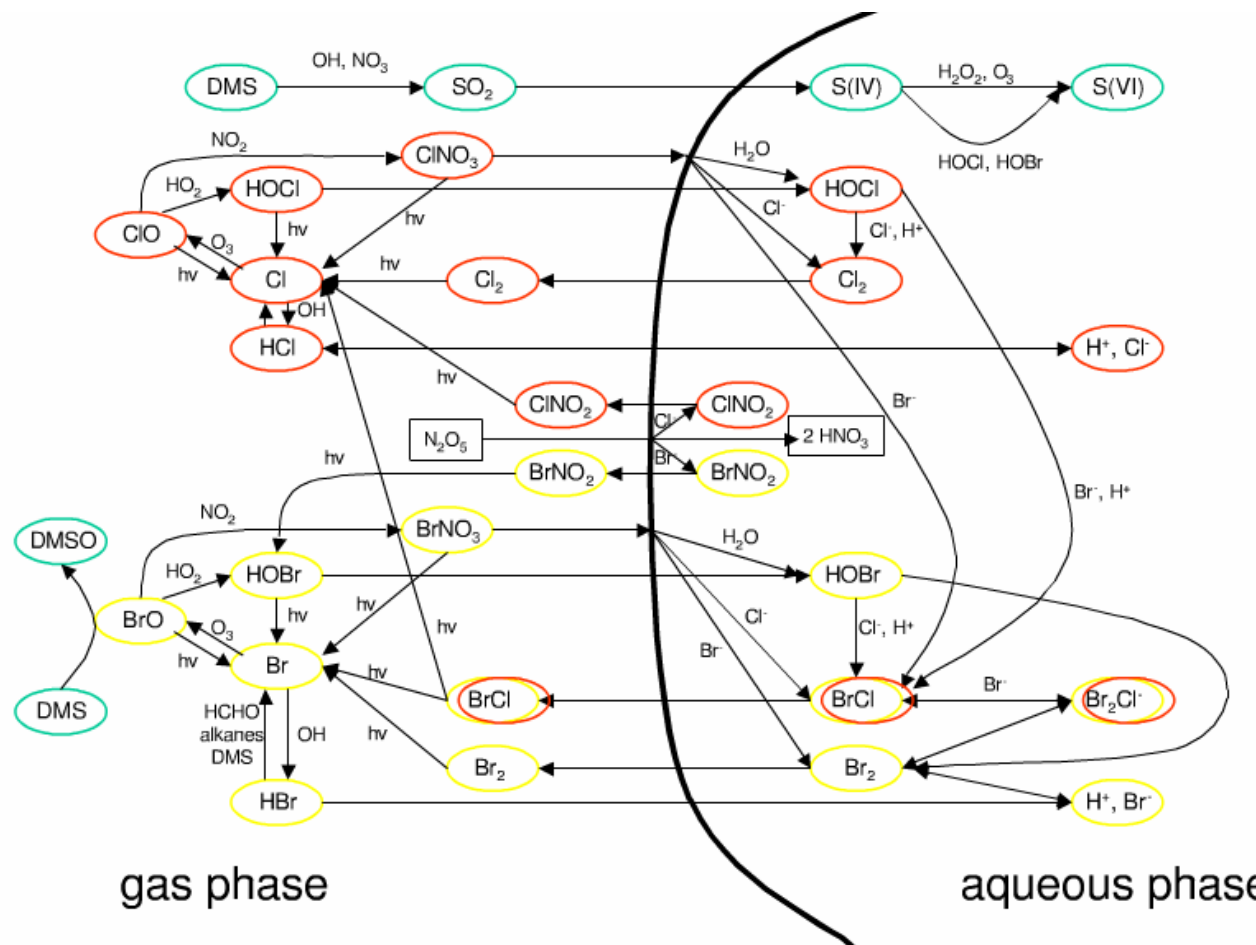
Abbatt et al., 2012

# To understand the ODEs



- We need to understand issues for each sub-field

# Issues for observations



Simpson et al., 2007

- Schematic diagram of the major halogen-related reactions

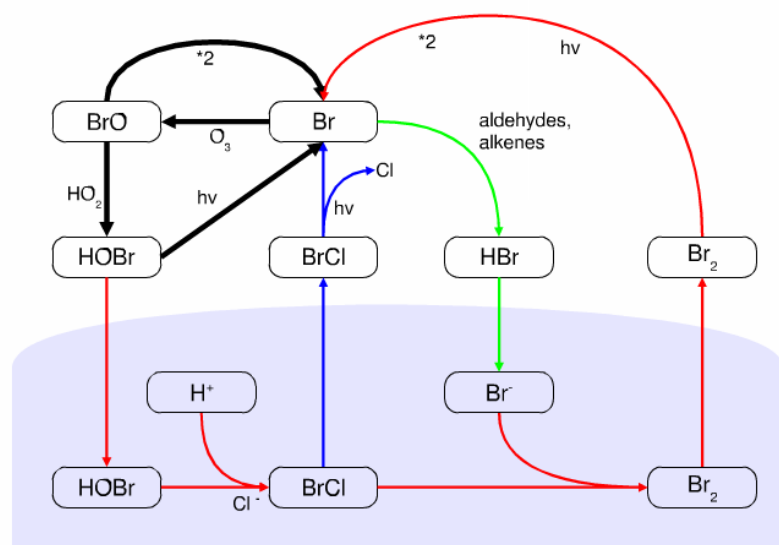
# Issues for observations

Examples of field observations ...

Instrument	Species/products/pros	Limits
Ozonesonde	O <sub>3</sub> profile (surface - ~30 km)	Temporal coverage (eg. 2/day)
UV photometric monitor	In-situ ozone measurements in snow and surface	Spatial coverage (eg. limited by tower height)
Ultraviolet differential absorption lidar (DIAL)	O <sub>3</sub> profile (few km - )	Depend on setup, may have low sensitivity in boundary layer/ limited by weather condition ...
MAX-DOAS	BrO profile	Low vertical resolution (optimal estimation, DOF ~ 2)
Longpath-DOAS	BrO, IO, and ClO	Depend on setup, may not provide vertical profile
Chemical ionization mass spectrometer	Br <sub>2</sub> , HOBr, BrCl, and Cl <sub>2</sub>	Spatial coverage
Sun photometer	AOD, solar flux	-
UV Satellite	O <sub>3</sub> , BrO, and IO	No profile information, need caution when derive VCD <sub>trop</sub>

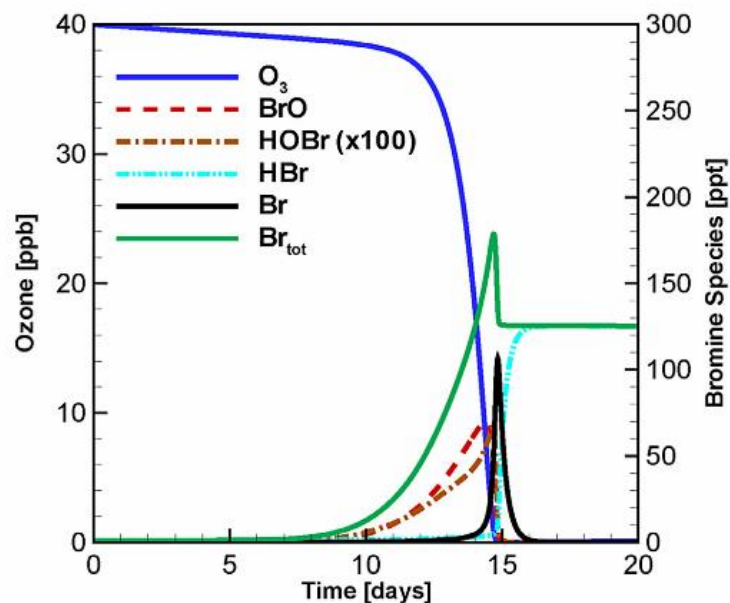
- Only a small fraction of the halogen compounds thought to be involved in the chain reactions has been detected in field measurements

# Issues for observations



**Fig. 4.** A simplified set of bromine explosion reactions. The blue area at the bottom is meant to represent the condensed phase (liquid brine or ice surface).

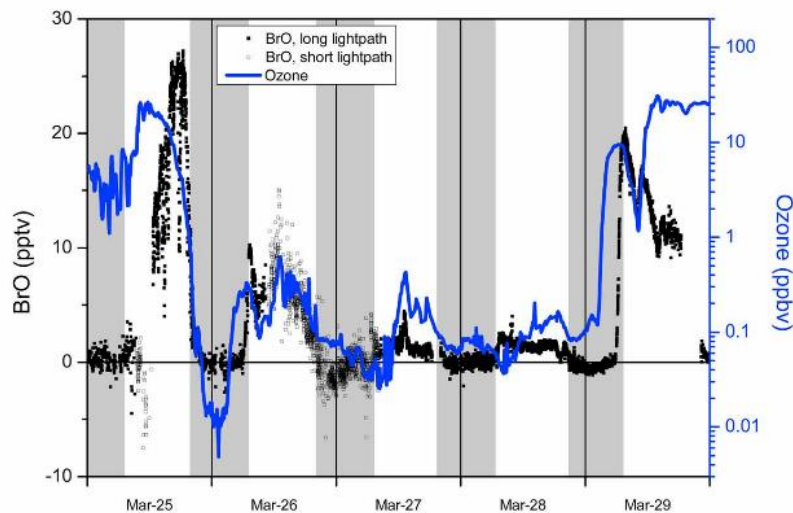
Simpson et al., 2007



**Fig. 4.** Temporal evolution of the mixing ratios of ozone and principal bromine species in the lowest layer of the model for a potential temperature gradient of  $0.0007 \text{ K m}^{-1}$  and a wind speed of  $5 \text{ m s}^{-1}$  (boundary layer height  $L = 200 \text{ m}$ ).

Cao et al., 2016

- Field measurements often only give brief observations of the chemical composition with a poorly determined history of the air mass (Simpson et al., 2007). One benefit of models is that the whole life cycle of ODEs and the shift in elemental speciation and oxidants can be investigated.

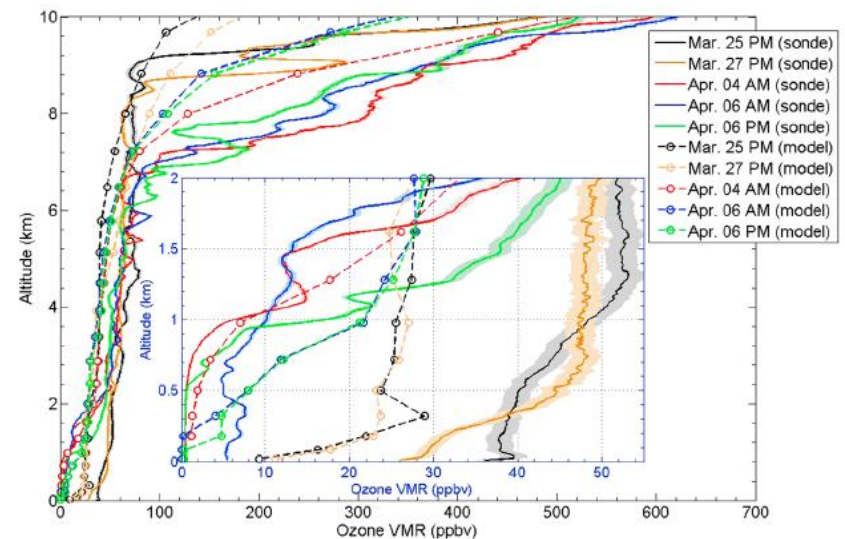


**Figure 7.** Surface ozone measured with the INSTAAR ozone chemiluminescence instrument and BrO mixing ratios measured by LP-DOAS from March 25–29. Open and closed black symbols indicate BrO measurements (scale on left axis) performed along the short and long light path, respectively. The ozone surface mixing ratio (blue line) is shown on logarithmic scale (right axis). Nighttime periods are shaded in gray.

Helmig et al., 2012

- Need observations of other bromide and chloride species in ODEs in sub-ppbv ozone condition

Zhao et al., 2016



**Figure 11.** Tropospheric ozone profiles above Eureka (the inset panel shows 0–2 km). Solid lines (as indicated in the legend) show ozonesonde measurements on 25 March (23:15 UTC), 27 March (23:15 UTC), 4 April (6:52 UTC), and 6 April (3:42 and 20:00 UTC) 2011, the shaded regions represent the 1 sigma uncertainty envelope. The dashed lines with circles show the UM-UKCA modeled ozone profiles for 25 March (23:00 UTC), 27 March (23:00 UTC), 4 April (7:00 UTC), and 6 April (4:00 and 20:00 UTC) 2011.

# Issues for models

- Need more constraints from observation
  - Only a small fraction of the halogen compounds thought to be involved in the chain reactions has been detected in field measurements to date
- Need more measurement data for evaluation
- Missing data on reaction kinetics, in particular the temperature dependence of some key reactions
- Need more detailed description of processes in and on the snow (and aerosol)
- Representation of aerosols
- Representation of the very shallow polar boundary layer

# Issues for lab

- Need to understand chemistry above ice/snow surfaces
- Need to understand the microstructural distribution of impurities on ice surfaces
- Many of the rate constants used in modeling of the halogen chemistry need better measurements and better definition of their temperature dependence
- Methods need to be developed to determine in-situ properties of ice surfaces, such as the effective pH of ice surfaces

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Atmospheric  
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## **A review of air–ice chemical and physical interactions (AICI): liquids, quasi-liquids, and solids in snow**

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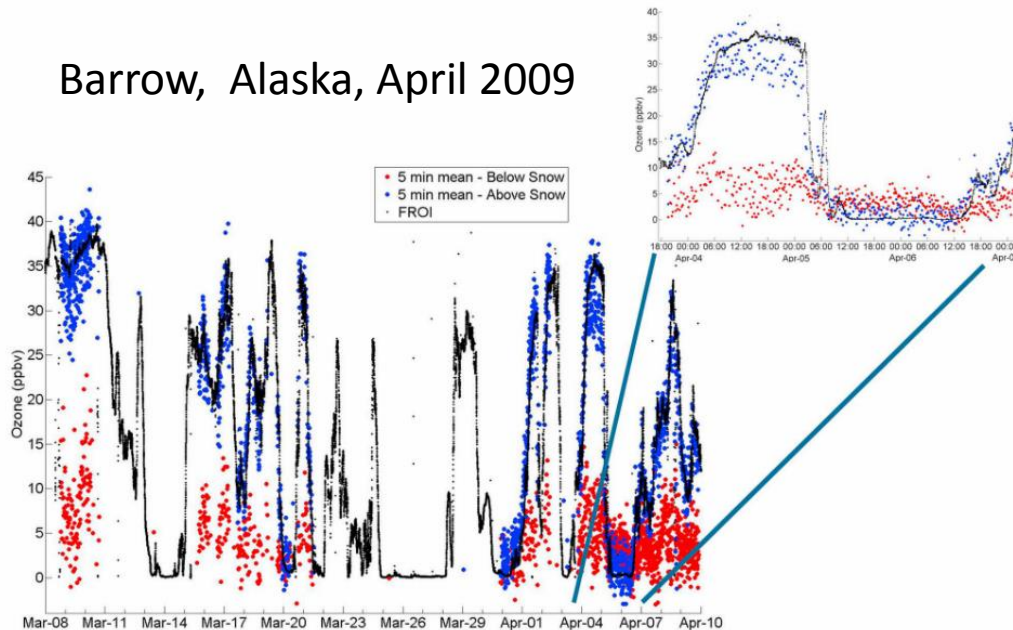
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# Outline

1. Brief review of surface ODEs & issues for lab, model, and observation
2. Few highlights of recent case studies
  - Ozone in the snow (model vs. measurements)
  - Partitioning halogen species (model constrained by measurements)
  - Modeled and MAX-DOAS BrO profile comparison, blowing snow & shallow boundary layer
3. Challenges & opportunities for observation group

Barrow, Alaska, April 2009



**Figure 1.** Sub-snow surface (−35 cm, red points) and above surface (+1 m, blue points) ozone mixing ratios from the switched inlet ozone experiment. Ambient air results from the FROI, shown in black a included for comparison.

#### 4. Summary and Conclusions

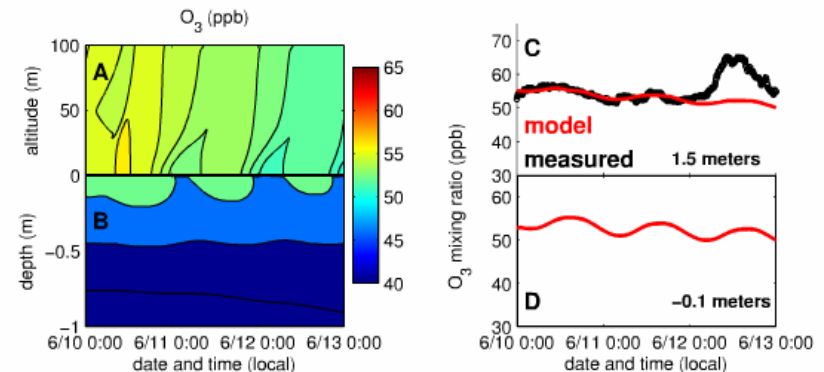
Helmig et al., 2012

[35] The ozone concentration and flux data from the multiple experimental platforms provide a number of new insights into ozone chemical and dynamical behavior in the coastal arctic environment. The snow covered tundra was found to be a sink of ozone. Ozone in snowpack air was depleted of ozone regardless of surface concentrations. There was no indication that ozone chemistry occurring in the snowpack at this site plays a significant role in determining the ODE dynamics seen above the surface. Ozone surface deposition rates were relatively low, on the order of  $\leq 0.02\text{--}0.05 \text{ cm s}^{-1}$  during most times, which is of similar order as seen at other snow covered arctic sites. There was no clear evidence of ozone in interstitial air being influenced by photochemical processes; ozone in the snowpack did neither show increases (from production) nor decreases (from chemical depletion) associated with diurnal radiation cycles. This behavior is in contrast to the snowpack ozone chemistry at Summit. Ozone production chemistry driven by

## Example cases

- Ozone in the snow during ODE

Summit, Greenland, June 2008



**Fig. 9.** Modeled  $\text{O}_3$  mixing ratios in the atmosphere (A) and interstitial (B) air. Modeled mixing ratios in the atmosphere at an altitude of 1.5 m above the snowpack are compared with measurements in (C). Predicted interstitial air mixing ratios 10 cm below the snow surface are shown in (D).

Thomas et al., 2011

# Example cases

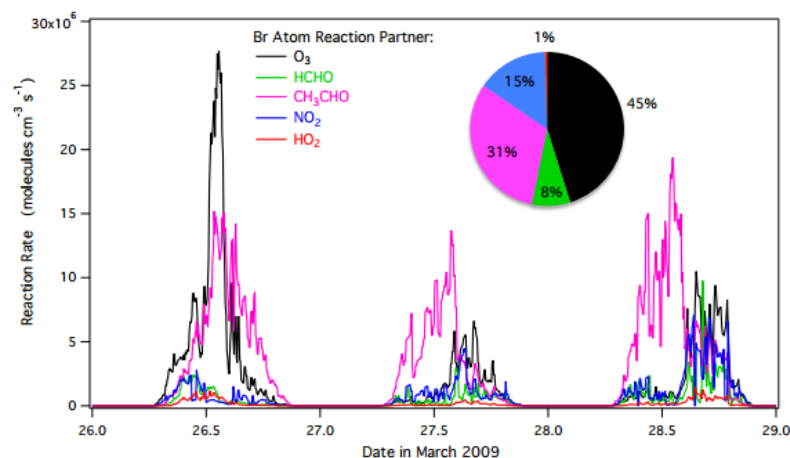
**Table 5.** Summary of the ambient measurements from OASIS that were used to constrain the model and the instrumental method used.

Measured species	Method	Reference
O <sub>3</sub> , NO, NO <sub>2</sub>	Chemiluminescence	Ridley et al. (1992), Ryerson et al. (2000)
CO	IR absorption CO analyzer	
Cl <sub>2</sub> , Br <sub>2</sub> , BrO, HOBr	Chemical ionization mass spectrometry (CIMS)	Liao et al. (2011, 2012, 2014)
ClO	Chemical reaction GC-ECD	Stephens et al. (2012)
HCHO	Tunable diode laser absorption spectroscopy	Fried et al. (1997), Lancaster et al. (2000)
HONO	Long-path absorption photometer	Villena et al. (2011)
CH <sub>3</sub> CHO, CH <sub>3</sub> COCH <sub>3</sub> , MEK, <i>n</i> -C <sub>4</sub> H <sub>10</sub> , <i>i</i> -C <sub>4</sub> H <sub>10</sub>	Online fast GC-MS	Apel et al. (2002)
C <sub>2</sub> H <sub>2</sub> , C <sub>2</sub> H <sub>4</sub> , C <sub>2</sub> H <sub>6</sub> , C <sub>3</sub> H <sub>8</sub> , C <sub>3</sub> H <sub>6</sub> , <i>n</i> -C <sub>4</sub> H <sub>10</sub> , <i>i</i> -C <sub>4</sub> H <sub>10</sub>	Canister samples, offline GC-MS	Russo et al. (2005)

- Partitioning halogen species (model constrained by measurements)

## 4 Conclusions

The goal of this work was to investigate the interactions and impacts of halogen chemistry on ozone depletion using a model that is constrained to a unique set of observed, time-varying chemical conditions at the time of the event. With this approach, we have been able to dissect some of the important chemical pathways pertaining to ODEs, focusing on the interactions between the halogen radicals. It is clear that the interactions between bromine, chlorine, and iodine are very complex and highly dependent on the concurrent conditions of relevant species, such as O<sub>3</sub>, HO<sub>x</sub>, NO<sub>x</sub>, and the VOCs. As these species fluctuate, the partitioning of halogen species will also change, and so too will their impact on chemistry of the PBL. Thus, a full understanding of halogen chemistry requires the careful measurement of all these species (including Cl<sub>2</sub>, Br<sub>2</sub>, HOBr, and HOCl).



**Figure 7.** Time-varying rates (in molecules cm<sup>-3</sup> s<sup>-1</sup>) of the important Br atom sinks during ODEs (O<sub>3</sub> < 5 ppbv). The inset pie chart represents the fractional importance of each sink calculated as the median value across the 3 days shown. Time is expressed in Alaska Standard Time.

# Example cases

elevated BrO often coincided with enhanced aerosol optical depth, and that there were sometimes enhanced layers of BrO and aerosol extinction. However, enhanced BrO and aerosol extinction were also sometimes close to the surface. Two example days are shown in Fig. 8, which shows AOD and BrO vertical profile data retrieved from the MAXDOAS, for 11 and 14 April 2009 at Barrow. Those observations imply that indeed activation of bromine on aerosol surfaces can be an important mechanism, e.g. as shown for 14 April. However, even for 11 April 2009, when the enhanced BrO is surface-based, that enhanced BrO was well-correlated with AOD. These observations point out the need for 1-D profiles of the halogen atom precursors ( $\text{Br}_2$ ,  $\text{BrCl}$ , and  $\text{Cl}_2$ ) along with aerosol size, number and composition data. The relationships between blowing snow and halogen activation have not been directly observed, during or after blowing snow events. In the

future, the autonomous monitors such as (Knepp et al., 2010; Bauguitte et al., 2011) will provide useful data to confirm if these events occur. In particular, it is important to be able to measure ozone, BrO and meteorological data simultaneously (Knepp et al., 2010).

During the OASIS2009 campaign, Liao et al. (2011b, 2012) conducted measurements of bromine species, including  $\text{Br}_2$ , HOBr, and BrO, via chemical ionization mass spectrometry (CIMS), obtaining maximum values of 45, 25 and 40 pptv, respectively. The HOBr and  $\text{Br}_2$  data are consistent with the Fan and Jacob (1992) mechanism, in that HOBr was observed with a diurnal profile consistent with daytime photochemical production, and night-time uptake on surfaces with simultaneous production of  $\text{Br}_2$ . Buys et al. (2012) report high resolution observations of  $\text{Br}_2$ ,  $\text{BrCl}$  and BrO made in coastal Antarctica using CIMS during Austral spring in

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Abbatt et al., 2012

- Modeled and measured BrO profile comparison
- Blowing snow & shallow boundary layer

# Example cases

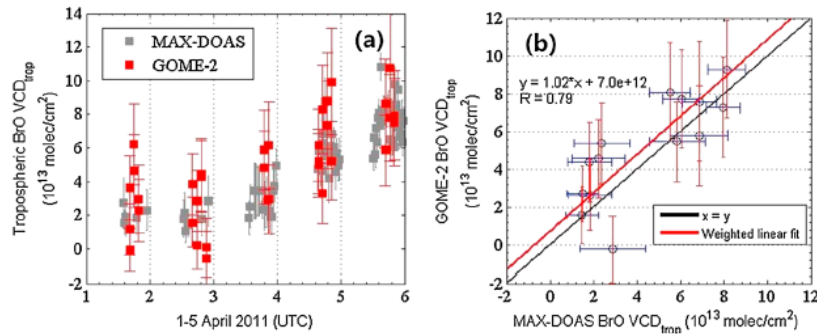
## A case study of a transported bromine explosion event in the Canadian high arctic

X. Zhao<sup>1</sup>, K. Strong<sup>1</sup>, C. Adams<sup>1,2</sup>, R. Schofield<sup>3,4</sup>, X. Yang<sup>5</sup>, A. Richter<sup>6</sup>, U. Friess<sup>7</sup>, A.-M. Blechschmidt<sup>6</sup>, and J.-H. Koo<sup>1</sup>

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### Key Points:

- A transported bromine explosion event was observed in the Canadian High Arctic
- Both blowing snow and a stable shallow boundary layer were observed during the event
- The bromine explosion event is simulated by a global chemistry-climate model



**Figure 6.** (a) Time series of BrO tropospheric partial column densities (VCD<sub>top</sub>) from PEARL-GBS MAX-DOAS (0–4 km altitude) and GOME-2 (approximately 0–8 km altitude, full troposphere) within 45 km of Eureka. (b) GOME-2 versus MAX-DOAS tropospheric BrO partial column densities. MAX-DOAS error bars are total retrieval error, based on *Frieß et al.* [2011]. GOME-2 error bars are systematic error, estimated based on *Theys et al.* [2011].

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## An exemplary case of a bromine explosion event linked to cyclone development in the Arctic

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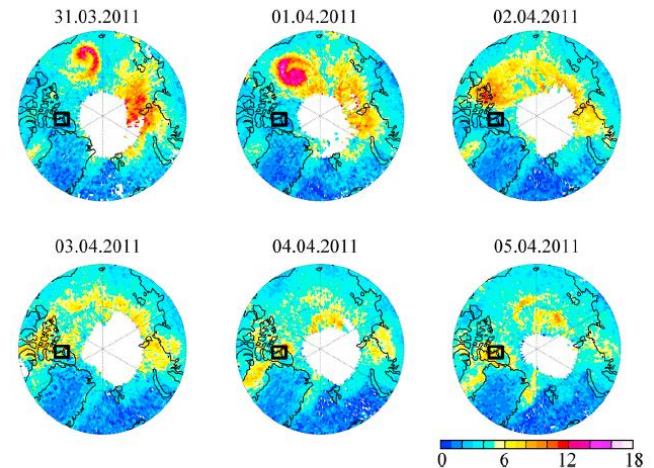
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<sup>3</sup>Department of Physics, University of Toronto, Toronto, Ontario, Canada

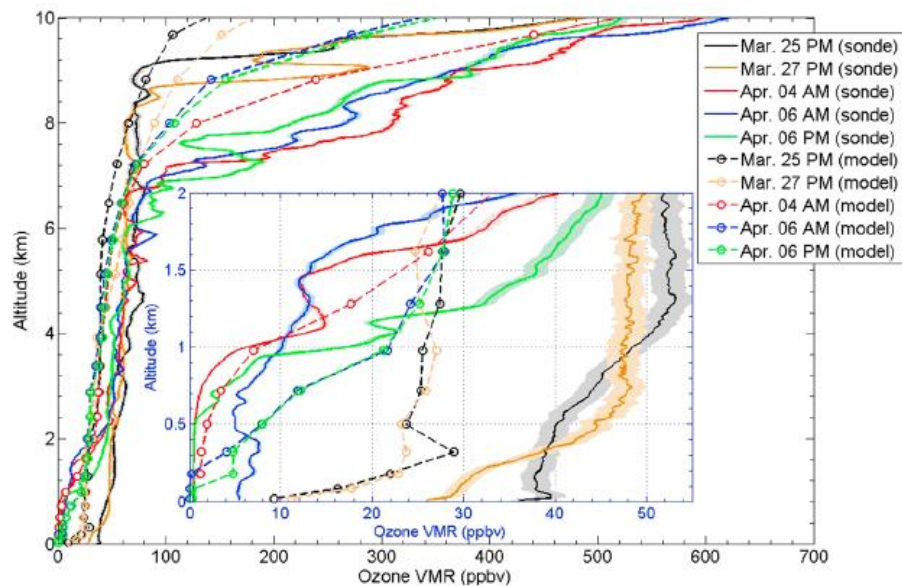
<sup>4</sup>Belgian Institute for Space Aeronomy (IASB-BIRA), Brussels, Belgium

<sup>a</sup>now at: Energy & Meteo Systems GmbH, Oldenburg, Germany



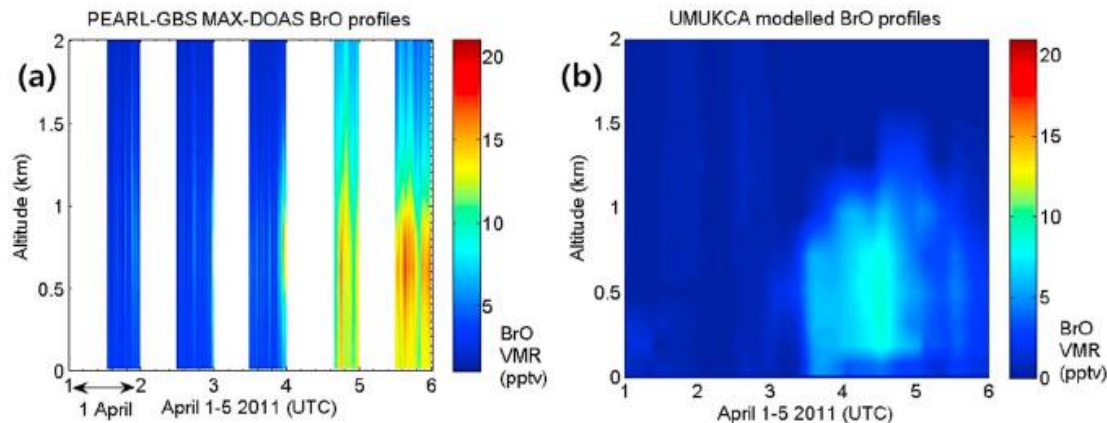
**Figure 8.** GOME-2 BrO VCD<sub>top</sub> (x10<sup>13</sup> molecules cm<sup>-2</sup>) north of 70°N from 28 March to 5 April 2011. The location of Eureka is indicated by the black square on each panel.

- Long-distance transportation (~1800km over 5 days) to Eureka indicated strong recycling of BrO within the bromine plume.



**Figure 11.** Tropospheric ozone profiles above Eureka (the inset panel shows 0–2 km). Solid lines (as indicated in the legend) show ozonesonde measurements on 25 March (23:15 UTC), 27 March (23:15 UTC), 4 April (6:52 UTC), and 6 April (3:42 and 20:00 UTC) 2011, the shaded regions represent the 1 sigma uncertainty envelope. The dashed lines with circles show the UM-UKCA modeled ozone profiles for 25 March (23:00 UTC), 27 March (23:00 UTC), 4 April (7:00 UTC), and 6 April (4:00 and 20:00 UTC) 2011.

- Observation of surface ODEs (with elevated BrO) in both blowing snow and stable shallow boundary condition

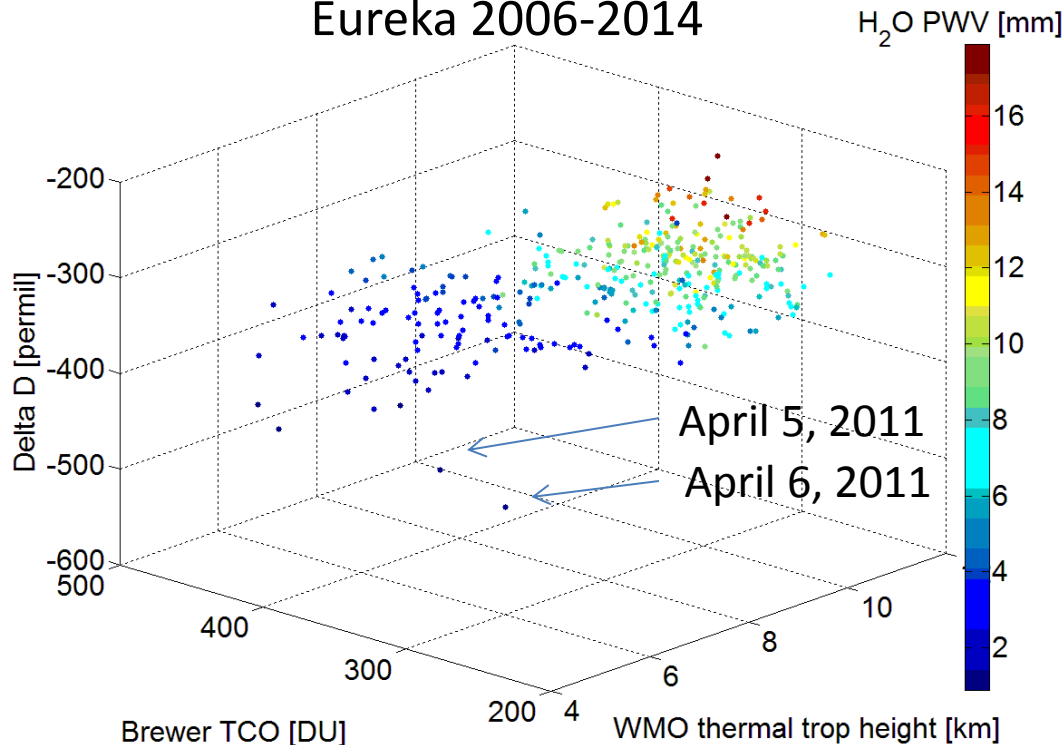


**Figure 15.** (a) MAX-DOAS tropospheric BrO profiles measured at Eureka; (b) UM-UKCA modeled tropospheric BrO profiles over Eureka on the same color scale as Figure 15a. Note that the modeled BrO in Figure 15b is the same as in Figure 14b. Figure 15 highlights the lowest 2 km and uses a different color scale for comparison with the MAX-DOAS measurements.

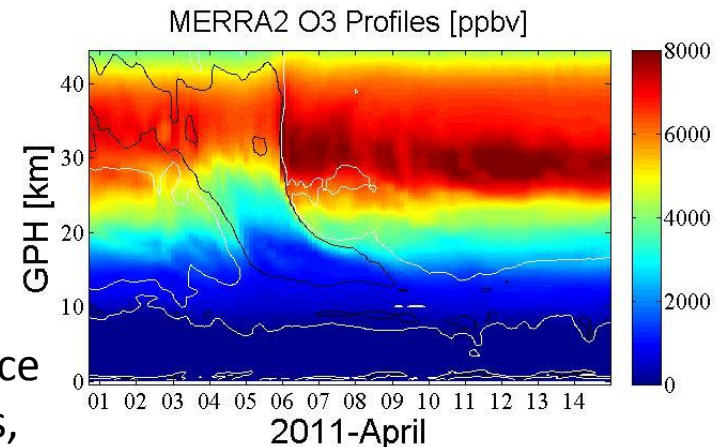
- The chemistry-climate model successfully reproduced some of the main features (such as the vertical structure) of this BEE after including a bromine source from blowing snow related to sea-salt production.

# Example cases

O<sub>3</sub> + H<sub>2</sub>O + Delta D + trop heights at Eureka 2006-2014



- Surface O<sub>3</sub> depleted
- Very low stratospheric O<sub>3</sub> (polar vortex was above the site) → increased UV radiation
- Not an stratospheric intrusion
- HDO depleted → ice clouds/ ice partials
- Low H<sub>2</sub>O



- Measurements of HDO also contain information about ice cloud formation and evaporation. When cloud ice forms, HDO is concentrated in the ice and the air is HDO depleted.

**Preliminary work**

# Outline

1. Brief review of surface ODEs
2. Few highlights of recent case studies
3. Challenges & opportunities for observation group
  - re-address some open questions related to observations
  - challenges & opportunities for observation groups

# Open questions

Following the two review paper

- Substrates for halogen activation
  - First-year sea ice
  - Frost flower
  - Salty snow (ground and lofted)
  - Aerosol
- Active halogen sources
  - Inorganic vs. organic sources
- Role of meteorology in boundary layer ozone loss
  - Initialization of ODEs
  - Transportation of ODEs
  - Termination of ODEs
- Effects of climate change

# Arctic Ozone - Key Science Questions

1. Are we measuring ozone with sufficient accuracy, geographical coverage and vertical resolution? (see also question 10)
2. Why does the seasonal cycle vary across the Arctic? What are the processes that affect seasonal cycle? Can we identify how it has changed over time from long-term measurements?
3. What is the importance of classical NO<sub>x</sub>, VOC chemistry compared to halogen chemistry?
4. Does ozone in the Arctic increase?
5. Can we link changes in Arctic ozone to climate change?
6. Are Arctic ozone changes influenced by the tropical and middle latitude emissions of precursors? Can we identify the origins of these sources by using complementary and colocated measurements of carbon isotopes in the Arctic?
7. Do forest fires impact surface ozone levels in the Arctic?
8. How and where geographically do long transports of air masses mix into the boundary layer? Are there deposition processes that impact ozone in air masses during transport?
9. How well can global transport and chemistry climate models simulate seasonal and intra-annual variability in ozone within and above the boundary layer?
10. What is the vertical distribution of ozone concentrations up through the troposphere?
11. What do we need before we can formulate parameterization for ESM to catch the variability of ozone in Arctic?

# Challenges & opportunities for observation groups

- Need measurements of more species (Br, HOBr, IO, Hg, and etc ...)
  - With only ozone (and BrO) measurements, we could not retrieve the big-picture of surface ODEs
- Need improved vertical profiles
  - Ozone and halogen measurements in snow layers
  - Ozone and halogen measurements in UT-LS
- Need improved spatial/temporal coverage
- Need detailed meteorological measurements
  - Clouds condition/type
  - Aerosol type
  - UV radiation
  - Water vapor

# Possible future topics

- Conditions for the termination of ODEs
- Link ozone vertical flux with halogen vertical flux
- Long-term measurements for atmospheric mercury depletion events (AMDEs)
- Organic halogen source
- ...

A photograph of a snowy mountain landscape. In the foreground, a snow-covered railing with yellow-painted metal posts runs diagonally across the frame. To the right, a small, clear, dome-shaped structure sits on a platform. The background shows a vast, snow-covered mountain range under a bright, hazy sky with a large, glowing sun. The overall scene is serene and cold.

# Thank You

Picture of a blowing snow event at Eureka